

1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22

Trade-offs between nutrient circularity and environmental impacts in the management of organic waste

Selene Cobo*, Antonio Dominguez-Ramos and Angel Irabien

Department of Chemical and Biomolecular Engineering, University of Cantabria

Avda. los Castros s.n., Santander, 39005, Spain

*Corresponding author: Selene Cobo

Tel.: +34 942 20 09 31. E-mail: cobos@unican.es

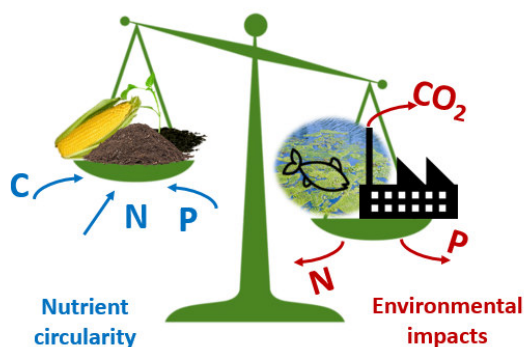
23 **ABSTRACT**

24

25 Measuring the circularity of resources is essential to assess the performance of a circular
26 economy. This work aims at proposing an indicator that quantifies how effective a system is at
27 extending the lifetime of its waste components after they have been discarded. The developed
28 indicator was applied to study the circularity of nutrients within a system that handles the
29 organic waste (OW) generated in the Spanish region of Cantabria. A superstructure was
30 developed to determine the optimal configuration of the system. It comprises alternative Unit
31 Processes (UPs) for i) the management of OW, and ii) the application of the recovered products
32 as soil amendment to grow corn. A multi-objective Mixed Integer Linear Programming problem
33 was formulated under two policy scenarios with different source separation rates (SSRs). The
34 problem was optimized according to six objective functions: the circularity indicators of carbon,
35 nitrogen and phosphorus, which are maximized, and their associated environmental impacts to
36 be minimized (global warming, marine eutrophication and freshwater eutrophication). The
37 model was fed with the Life Cycle Assessment results obtained with EASETECH (Environmental
38 Assessment System for Environmental TECHNOlogies) and the nutrient flows in the agriculture
39 subsystem, which were calculated with DNDC (DeNitrification-DeComposition). It was
40 concluded that improving nutrient circularity paradoxically leads to eutrophication impacts, and
41 increasing the SSR of OW has a positive effect on the carbon footprint of the system.

42

43 **Table Of Contents**



44

45 **INTRODUCTION**

46

47 In the context of a boom of initiatives promoting a circular economy within the European
48 Union,¹⁻³ it is the responsibility of researchers to provide policy-makers with the data and tools
49 needed to make informed decisions. Measuring the circularity of resources is key to assessing
50 the performance of a circular economy.

51

52 **Literature overview**

53 Several approaches have been presented to tackle this challenge. One study defined a global
54 circularity indicator as the share of material inputs into the global economy that are cycled,
55 subsequently estimating that the global economy was 9.1% circular in 2015.⁴ Although this
56 indicator provides insight into the global materials metabolism, policy implications cannot be
57 directly derived from it. Instead, an indicator that can be applied to systems design and
58 operation is of more interest to the policy makers.

59

60 Some authors suggest that circularity indicators should capture how the differences between
61 the physico-chemical properties of the recovered waste components and the primary resources
62 they displace affect their substitution ratio.⁴⁻⁷ Accordingly, Moriguchi⁵ pointed out that the
63 reduction in the requirement for primary resources could be a good indicator of circularity.
64 However, this does not necessarily entail that more waste components are being recovered; it
65 could be the consequence of an increase in the eco-efficiency of the system.

66

67 Haupt et al.⁶ suggested that open-loop and closed-loop recycling rates that reflect the efficiency
68 of the recycling processes and the type of application of the recycled components in their next
69 life cycle stage should be used as performance indicators for a circular economy.

70

71 The duration of material retention within a system has also been recommended as an indicator
72 of circularity.⁷ Following this line of thinking, the Ellen MacArthur Foundation proposed the
73 lifetime of a product as one of the parameters used to calculate its circularity indicator.⁸
74 Although this indicator is useful for companies, it does not provide information about the
75 circularity of the components of the product, since it does not consider their entire life cycle.

76

77 The described indicators do not correlate with the quality of the recovered components and
78 they do not reveal how much of the recovered components are consumed again; i.e., to what
79 extent the loop is closed.

80

81 The methodology proposed by Cobo et al.,⁹ which enables to track waste components within a
82 Circular Integrated Waste Management System (CIWMS), might help overcome these
83 limitations, since CIWMSs encompass not only waste management, but also the processing and
84 consumption of the components recovered from waste and the external raw materials that
85 eventually become waste.

86

87 **Case study**

88 This framework is applied to the study of the management of organic waste (OW) in the region
89 of Cantabria, in the north of Spain. The OW generated in Cantabria ($83.5 \cdot 10^3$ metric ton in 2014)
90 is collected with other discarded household inorganic materials. The OW that is sorted out at
91 the regional mechanical-biological treatment facility is subjected to a windrow composting
92 process. Nonetheless, Directive 2008/98/EC¹⁰ does not allow the land application of the bio-
93 stabilized material derived from the composting of the OW separated from the mixed waste
94 stream (mix-OW); only the OW that has been source separated (SS-OW) can be recycled. The
95 expiration of the regional authorization that permitted the sale of the bio-stabilized material as
96 compost until 2018¹¹ makes it impossible for the current waste management system to comply

97 with the legal restraints. The need to retrofit the system represents an opportunity to
98 implement new circularity practices. The interest of recycling OW lies in the nutrients it contains.
99

100 **Implications of nutrient recovery**

101 This study focuses on three essential nutrients to soil amendment: carbon (C), nitrogen (N) and
102 phosphorus (P). Enhancing the circularity of these nutrients within a CIWMS *a priori* seems to
103 be a strategy that will contribute to closing their natural biogeochemical cycles by avoiding the
104 accumulation of nutrients in one of the Earth's subsystems (atmosphere, hydrosphere,
105 biosphere or lithosphere) at a rate faster than the ecosystems can sustain. Thus, the relevance
106 that a circular economy of nutrients might have to global sustainability challenges should not be
107 underestimated. On the one hand, the forthcoming peak P production, due to the depletion of
108 the global rock phosphate reserves, threatens future food security;¹² on the other, the
109 anthropogenic interference with the C and N biogeochemical cycles to meet the energy and food
110 demands has already caused the transgression of the estimated climate change and N cycle
111 planetary boundaries within which humanity is expected to operate safely.¹³

112

113 Since the nutrient cycles interact with each other,¹⁴ promoting the circularity of one nutrient
114 might have consequences on the biogeochemical cycles of the others. For instance, increasing
115 Soil Organic Carbon (SOC) stocks may exacerbate N₂O emissions,¹⁵ and an increased availability
116 of reactive N may lead to C sequestration because of biomass growth.¹⁶ Another counter-effect
117 related to the land application of the products recovered from OW is the accumulation of surplus
118 P in agricultural soils, because the N:P ratio in organic fertilizers is lower than the N:P ratio
119 required by crops.¹⁷⁻¹⁹

120

121

122

123 **Aim of the work**

124 The circularity of C, N and P within a CIWMS and the main impacts associated with the emissions
125 of these elements to the environment (global warming, marine eutrophication and freshwater
126 eutrophication) must be jointly analyzed. Although the recovery of nutrients is a subject that is
127 drawing the attention of the scientific community,²⁰⁻²⁴ the trade-offs between these indicators
128 have not been systematically explored in the literature yet. Therefore, the objectives of this
129 paper are the following:

- 130 - To propose a circularity indicator that can be applied to any non-renewable resource and
131 accounts for the extended service of the components recovered from waste.
- 132 - To optimize the OW management system in the region of Cantabria, setting as objective
133 functions the maximization of the circularity indicators of C, N and P, and the minimization
134 of the global warming, marine eutrophication and freshwater eutrophication impacts.

135

136

137 **METHODOLOGY**

138

139 Material Flow Analysis (MFA), Life Cycle Assessment (LCA) and multi-objective optimization
140 were applied to determine the optimal configuration of the Cantabrian CIWMS aiming at
141 nutrient recovery from OW. A superstructure comprising the combinations of unit processes
142 (UPs) that could emerge as a result of the optimization was proposed, as shown in Figure 1.
143 The UPs that already belong to the Cantabrian waste management system are represented with
144 a discontinuous line.

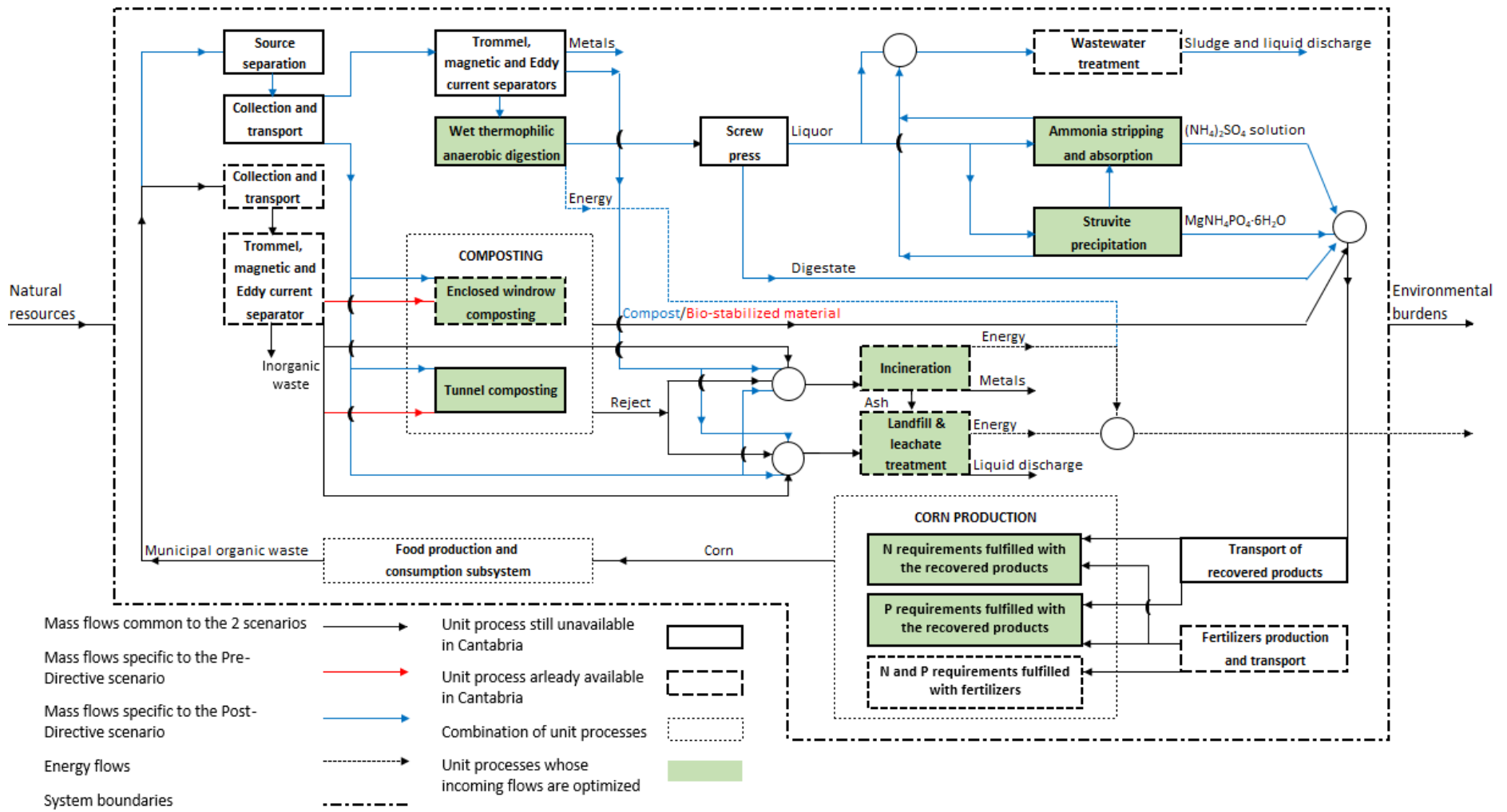


Figure 1. Studied CIWMS

145

146

147 **Superstructure description**

148 The products recovered from OW were assumed to be applied to land to grow corn, the main
149 fodder crop grown in Cantabria.²⁵ The superstructure comprises a set j of UPs for the
150 management of OW and a set k of corn production UPs. The UPs that can handle the solid OW
151 are wet thermophilic anaerobic digestion, windrow composting inside an enclosed building,
152 composting inside a tunnel reactor, incineration and landfill. The ammonia stripping and
153 absorption and the struvite precipitation UPs recover nutrients from the liquid digestate (LD)
154 produced in the anaerobic digestion, which only processes SS-OW after it has been pretreated.²⁶⁻
155 ³⁰ The remaining liquor is sent to a wastewater treatment plant. Incineration and landfill can also
156 handle the rejects generated by the other UPs. It is assumed that all the waste processing units
157 are in the same facility. A detailed description of these UPs can be found in Cobo et al.³¹

158

159 The nutrient uptake efficiencies of corn (shown in Appendix D of the Supporting Information)
160 differ for each type of applied product (bio-stabilized material, compost, digestate, struvite and
161 ammonium sulphate). As shown in Appendix C of the Supporting Information, P is in excess with
162 respect to the amount of N required by corn in all the recovered products except for ammonium
163 sulphate. Consequently, the nutrient flows were modeled so that the optimal approach to corn
164 production can be either based on one of these strategies or on a combination of them:

165 S1) Application of the amount of recovered product needed to cover the corn N requirements.

166 Unless ammonium sulphate is recovered, excess P is applied to soil, leading to freshwater
167 eutrophication.

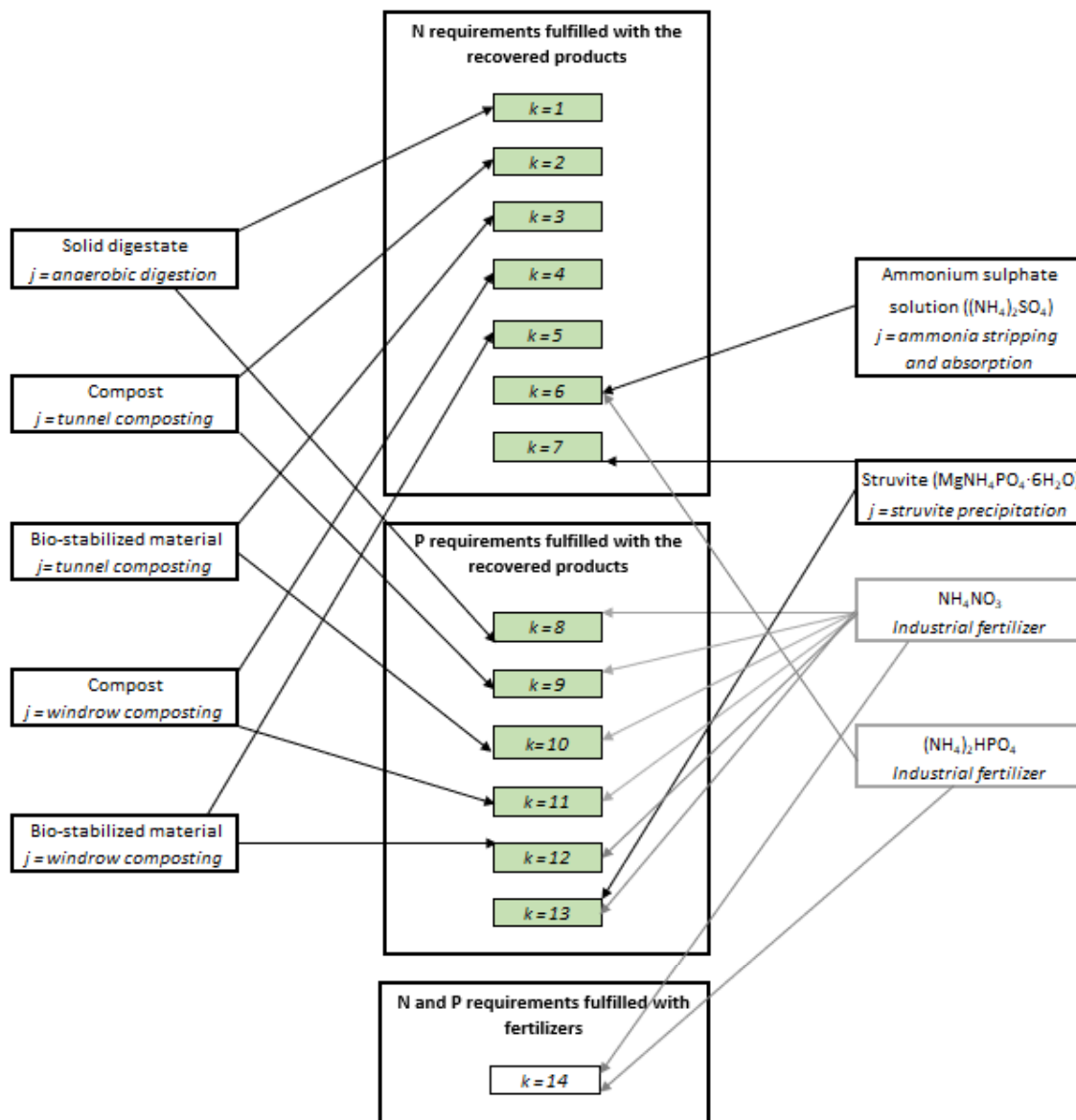
168 S2) Application of the amount of recovered product needed to cover the corn P requirements.

169 The N requirements are fulfilled with an industrial fertilizer (NH_4NO_3).

170 S3) Application of industrial N and P fertilizers (NH_4NO_3 and $(\text{NH}_4)_2\text{HPO}_4$).

171

172 The alternative combinations of the corn production UPs that can arise from the application of
 173 these strategies are shown in Figure 2. The N and P requirements of corn are defined as the
 174 amounts of these nutrients that yield the maximum average annual crop production that can be
 175 achieved in a 100-year timeframe with industrial N and P fertilizers. Assuming an 80% collection
 176 rate of the produced corn grain, it corresponds to a net production of 7.11 tons of corn grain per
 177 ha per year.



195 **Figure 2.** Possible combinations of inputs to the corn production subsystem

196
197
198

199 **Data flow**

200 A modular LCA approach, where the LCA of the individual UPs of the system is carried out,^{32,33}
201 was performed. The UPs concerning the management of solid OW were modeled with EASETECH
202 2.3.6,³⁴ which provided their environmental impacts. The nitrate and phosphate leachate, the
203 emissions of CO₂, N₂O and NO, the amount of Dissolved Organic Carbon (DOC) consumed by soil
204 microorganisms, the flows of N and P uptaken by corn and the amount of nutrients stored in soil
205 per hectare of cultivated corn were calculated with DNDC 9.5.³⁵ These results were transferred
206 to EASETECH 2.3.6, where the environmental impacts associated with the land application of
207 the recovered products and corn production were calculated.

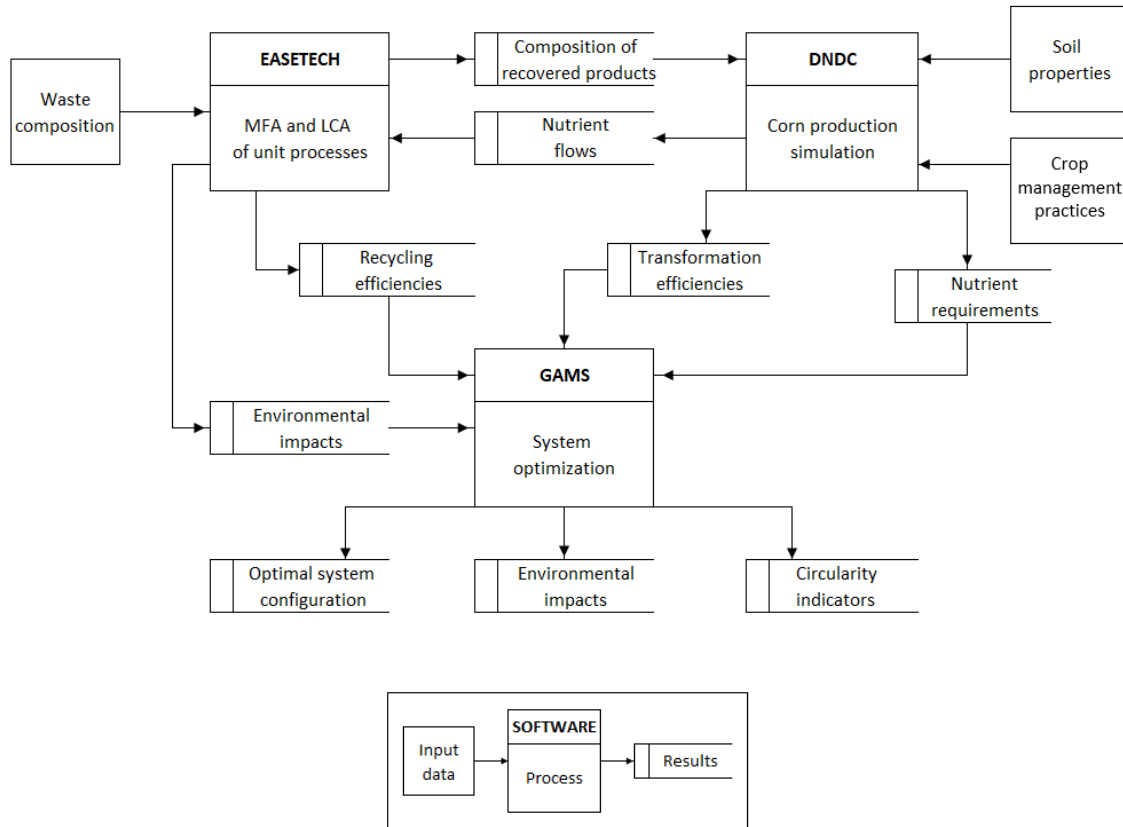
208

209 The results obtained with DNDC and EASETECH were exported as parameters to GAMS (General
210 Algebraic Modeling System) 24.8.1, where the problem was formulated. Figure 3 clarifies the
211 data flows derived from the application of this methodology.

212

213 The data required to characterize the UPs that integrate the system are compiled in the
214 Supporting Information: waste composition (Appendix A), waste management UPs (Appendix B)
215 and corn production subsystem (Appendix C).

216



217

218

219

Figure 3. Data flow diagram

220

221

222 DEFINITION OF THE CIRCULARITY INDICATORS

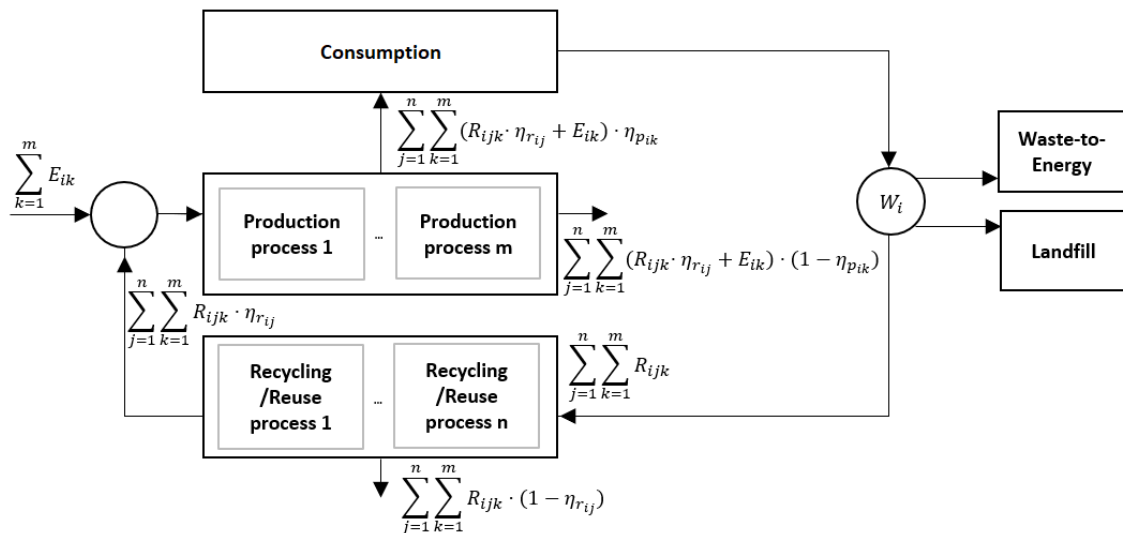
223

224 Figure 4 illustrates the flows of the component i of a given waste stream within a CIWMS. The
 225 circularity indicator of component i (CI_i) is defined as the amount of component i that extends
 226 its lifetime by providing a service in the upstream processes with respect to the amount of that
 227 component present in the collected waste. Equation 1 shows how the CI_i is calculated for a set
 228 of n recycling and preparation for reuse processes and m production processes that valorize this
 229 component.

$$CI_i = \frac{\sum_{k=1}^m \sum_{j=1}^n R_{ijk} \cdot \eta_{r_{ij}} \cdot \eta_{p_{ik}}}{W_i} \quad (1)$$

230 The variables needed for the calculation of CI_i are these:

- 231 - W_i . Amount of component i present in the waste stream (kg).
- 232 - R_{ijk} . Amount of component i that enters the recycling or preparation for reuse process
- 233 j . The subsequently recovered component i enters the production process k (kg).
- 234 - $\eta_{r_{ij}}$. Efficiency of the recycling or preparation for reuse process j for component i (kg of
- 235 component i recovered per kg of component i that enters process j).
- 236 - $\eta_{p_{ik}}$. Efficiency of the production process k at transforming or incorporating the
- 237 recovered component i into a product that will deliver a service in the consumption
- 238 subsystem (kg of component i transformed per kg of component i that enters process
- 239 k).



240

241

Figure 4. Simplified CIWMS

242

243 CI_i is dimensionless, its value can range between 0 and 1. A value of 1 implies that the total

244 amount of component i that was discarded is recovered and reprocessed to enter the

245 consumption subsystem, indicating that there are not any losses of component i in the recycling,

246 preparation for reuse and upstream processes. If $CI_i = 0$, component i is not recovered at all,

247 but incinerated or landfilled instead.

248

249 The proposed indicator indirectly accounts for the quality of the recovered components by
250 quantifying how much of the recovered component is consumed. This indicator does not
251 account *per se* for the degradation of the waste components after successive cycles, but if the
252 selected time horizon of the study is wide enough, a dynamic analysis should show how for a
253 sustained service demand, the external supply of component i ($\sum_{k=1}^m E_{ik}$) must increase due to
254 the degradation of the recovered component.

255

256

257 **Nutrient circularity indicators**

258

259 The circularity indicators of N and P (CI_N and CI_P) were defined as the amount of nutrient i that
260 is recycled, applied to land and uptaken by corn with respect to the amount of nutrient i present
261 in the collected OW.

262

263 The same definition cannot be applied to the C circularity indicator (CI_C), since the C captured
264 by vegetation in the photosynthesis process does not come from the soil but from the
265 atmosphere.

266

267 Besides improving the water-holding capacity of soil and its ability to retain cations in a plant
268 available form, contributing to C sequestration and promoting the formation of soil
269 structure,^{36,37} the purpose of applying a source of C to land is to feed the soil microorganisms.
270 When these microorganisms decompose the SOC, the decomposed C is partially lost as CO₂, and
271 DOC is produced as an intermediate that can be consumed by the soil microorganisms.³⁸ These
272 microbes are also responsible for the N fixation, ammonification and nitrification processes that
273 release N compounds that plants can assimilate; they are essential for crop production.

274

275 Consequently, a different definition was proposed for CI_C . It was defined as the ratio between
 276 the mass of DOC that is recycled, applied to land and consumed by microbes with respect to the
 277 amount of C present in the collected waste.

278

279 The values of $\eta_{r_{ij}}$ and $\eta_{p_{ik}}$ required for the calculation of the circularity indicators are compiled
 280 in Appendix D of the Supporting Information.

281

282

283 **PROBLEM FORMULATION**

284

285 A single-period Mixed Integer Linear Programming problem was formulated for the optimization
 286 of the decision variables; i.e., the incoming material flows (waste and recovered products) to
 287 the green shaded UPs in Figure 1. The problem was optimized according to these objective
 288 functions, where x and y represent the continuous and binary variables respectively: the
 289 circularity indicators of the studied nutrients, which must be maximized ($CI_C(x, y)$, $CI_N(x, y)$,
 290 and $CI_P(x, y)$), and the selected environmental impacts of the system to be minimized (global
 291 warming $GW(x, y)$, marine eutrophication $MEU(x, y)$ and freshwater eutrophication
 292 $FWE(x, y)$).

293

294 After verifying the trade-offs between the objective functions, a multi-objective problem was
 295 formulated as follows:

$$296 \min U(x, y) = \{GW(x, y), MEU(x, y), -CI_N(x, y), -CI_P(x, y)\} \text{ s. t. } \begin{cases} h(x, y) = 0 \\ g(x, y) \leq 0 \\ x \in \mathfrak{R}^n \\ y \in \{0, 1\}^m \end{cases} \quad (2)$$

297

298 The equations that describe the behavior of the system ($h(x, y) = 0$) are based on the mass
 299 balances of the UPs. The problem is subjected to these restrictions ($g(x, y) \leq 0$):

- 300 - The area fertilized with the recovered products cannot exceed the available area to grow
301 corn in Cantabria (4810 ha).³⁹
- 302 - The amount of biodegradable waste sent to landfill must be lower than 35% of the domestic
303 waste generated in 1995 (170,168 ton),¹¹ as established by Directive 1999/31/EC.⁴⁰
- 304 - Windrow and tunnel composting cannot accept waste streams with the same composition.
- 305 - SS-OW and mix-OW cannot be mixed in any composting processes.

306

307 The GAMS model comprises a total of 844 equations, 19 inequations, 817 continuous variables
308 and 28 discrete variables. The main input parameters to the models are the source separation
309 rate (SSR), the total area available for corn production and the amount of OW generated yearly
310 in Cantabria.

311

312 Different waste collection systems for SS-OW and commingled waste were modeled. It was
313 considered that the composition of SS-OW is 98% OW and 2% impurities, which is consistent
314 with documented source separation experiences.⁴¹ Two scenarios (neglecting and considering
315 the current legislative framework) were analyzed:

- 316 - **Pre-Directive scenario.** Mix-OW can be recycled. The SSR is 0% and no recycling target is set.
317 The red arrows in Figure 1 represent the flows of mix-OW that can only be composted in
318 this scenario.
- 319 - **Post-Directive scenario.** Mix-OW cannot be recycled. To comply with the 50% OW recycling
320 target established by the Cantabrian waste management plan¹¹ for 2020, a 50% SSR is set,
321 and an additional restriction is added to the model to ensure that 50% of the collected OW
322 is composted or anaerobically digested. The blue arrows in Figure 1 represent the flows of
323 SS-OW that are specific to this scenario.

324

325 The multi-objective optimization problem was solved with the CPLEX solver and the ϵ -constraint
326 method.⁴²

327

328

329 **MODELING APPROACH AND ASSUMPTIONS**

330

331 The boundary that separates the studied CIWMS from the ecosphere (which provides the
332 natural resources consumed by the system and a sink for the generated environmental burdens)
333 and the rest of the technosphere is depicted in Figure 1.

334

335 Although crops are managed by farmers under controlled conditions in the technosphere, they
336 produce natural biotic resources. Hence, the boundary between technosphere and ecosphere is
337 difficult to identify for agricultural soils.⁴³ One of the strategies recommended by Notarnicola et
338 al.⁴⁴ to overcome the limitations of considering agricultural soils as part of the technosphere, is
339 to include the impacts of crop production on soil. In this study the land application of the
340 recovered products and the production of corn were modeled as a UP. Although the system was
341 optimized for 1 year of operation, the selected 100-year time horizon enabled to account for
342 the loss of soil quality due to soil nutrient depletion caused by the production of consecutive
343 annual crops. The average annual corn production and emission rates in that timeframe were
344 considered.

345

346 Corn enters the food production and consumption subsystem, which comprises the upstream
347 processes that transform corn and the other food commodities consumed in Cantabria into OW.
348 It composes the background subsystem of the CIWMS because its configuration does not affect
349 the results of the study;⁴⁵ only the flows and the composition of its inputs and outputs (corn and
350 waste) that connect it to other UPs are calculated.

351

352 According to Cobo et al.,⁹ the primary function of CIWMSs is to recover waste components so
353 that their service life in the upstream processes can be extended. In this case study the elements
354 recovered from OW are used for land fertilization and soil conditioning. Since the studied CIWMS
355 encompasses the entire corn production of the region, the functional unit selected to perform
356 the LCA of the system is defined as the area available to grow corn in Cantabria (4810 ha).³⁹

357

358 An attributional LCA approach was applied. The electricity generated at incineration, anaerobic
359 digestion and landfill is considered the secondary system function. The direct substitution
360 method was applied by expanding the system boundaries to include the generation of electricity
361 from the Spanish grid mix. A 100% substitution ratio was assumed.

362

363 The characterization factors of each emission were calculated with the hierarchical 100-year
364 perspective of the ReCiPe 1.11 method. The assumptions made by the DNDC model about the
365 distribution of nutrients in the environment can be found in Li et al.⁴⁶ Following the rationale
366 explained by Cobo et al.,^{9,31} only the biogenic C present in animal and vegetable food waste
367 (which can i) leach into the water, ii) be emitted to the atmosphere, or iii) be stored either in the
368 landfill or the soil as a result of the land application of the recovered products, as shown in
369 Appendix C of the Supporting Information) was considered neutral. The CO₂ derived from the
370 decomposition of SOC was also quantified as fossil C.

371

372 Regarding the limitations of the model, the environmental impacts related to capital goods were
373 excluded from the analysis. Moreover, this work assumes that all the P is in mineral form and
374 accessible for plants. Studies have shown that most of the P in the products recovered from OW
375 is in mineral form, but not all of it.⁴⁷⁻⁵⁰

376

377 On the contrary, the mineralization of organic N is quantified by the DNDC biogeochemical
378 model. The organic/inorganic N ratio was assumed to be 93/7 for the compost and bio-stabilized
379 material,⁵⁰ and 62.96/37.04 for the solid digestate.⁵¹

380

381 The DNDC model assumes a 60% microbial efficiency to calculate the amount of C incorporated
382 into microbial biomass in amended soils, defined as the ratio of C assimilated into microbial
383 biomass to residue C released by decomposition.⁴⁶

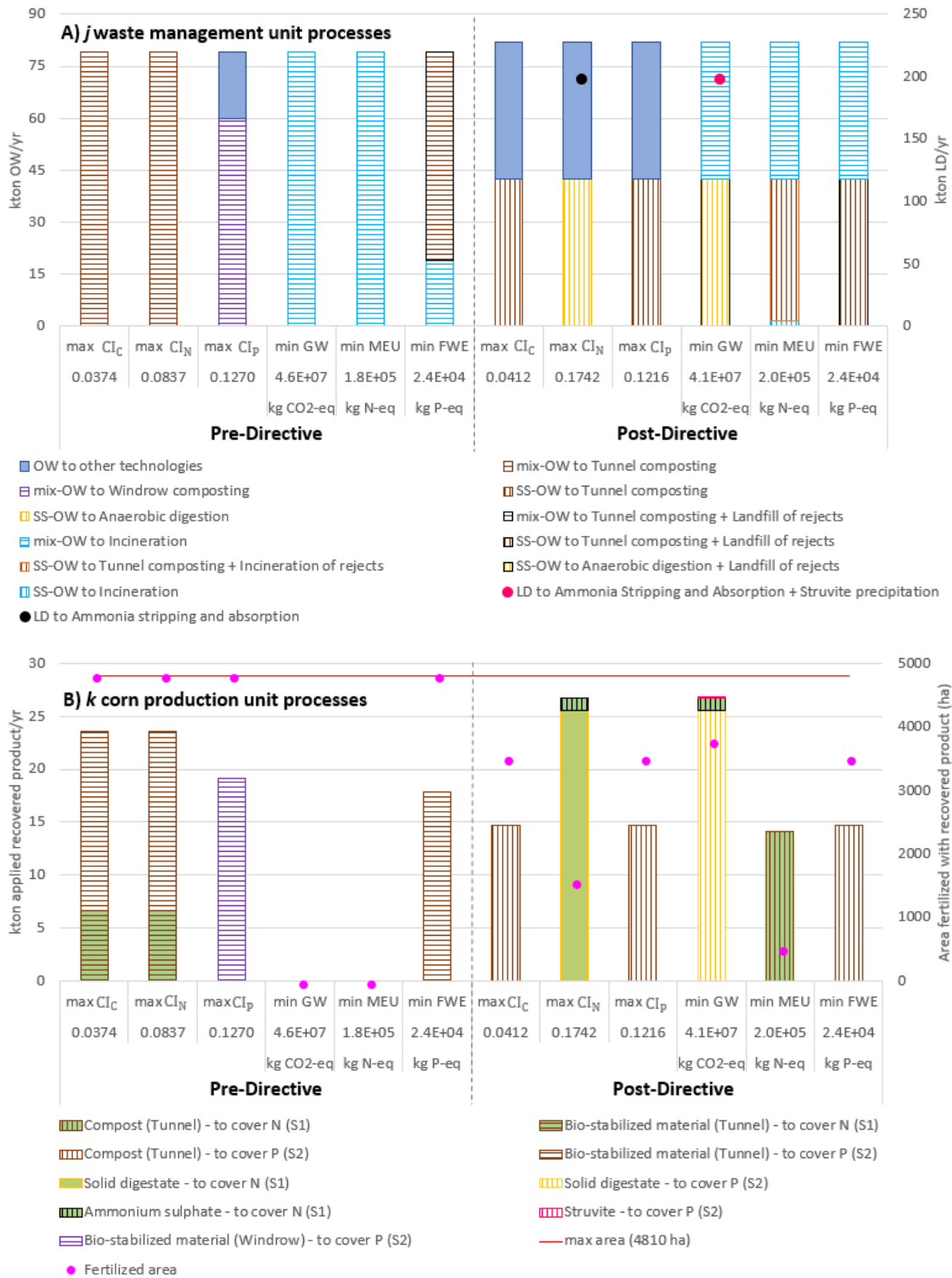
384

385

386 **RESULTS AND DISCUSSION**

387

388 The results of the problem optimization determine the system configuration; i.e., the UPs that
389 the system comprises and their incoming flows of waste and recovered products. The values of
390 the objective functions and the decision variables that optimize each objective function for the
391 two studied scenarios are compiled in Figure 5. Figure 5A shows the optimal flows of OW and
392 LD handled by the j UPs. The optimal flows of the recovered products into the k corn production
393 UPs (Figure 5B) are shown along with the area fertilized with the recovered products. The
394 contribution of the UPs to the environmental impacts of the optimal system configurations of
395 each scenario are depicted in Figure 6.



396

397

398 **Figure 5.** Values of the objective functions and decision variables for the optimization of the
 399 Pre-Directive and Post-Directive scenarios

400 The flows of OW shown in Figure 5A are lower in the Pre-Directive scenario because part of the
 401 OW present in the mixed waste ends up in the inorganic waste stream after the trommel
 402 separation required for the pretreatment of mixed waste.

403

404 There are several system configurations that lead to the maximization of a given circularity
405 indicator, because the UPs that manage the rejects do not affect the corn production subsystem,
406 and thus they do not contribute to closing the nutrient loops. By analogy, in the Post-Directive
407 scenario where mix-OW cannot be recycled, the selection of any UP for its management will
408 result in the same circularity indicators. This is the reason the maximization of the circularity
409 indicators in Figure 5A only shows the UPs that contribute to recirculate nutrients.

410

411 The amount of P present in the mix-OW collected in the Pre-Directive scenario is more than
412 enough to cover the P requirements of the corn produced in Cantabria under the hypothesis of
413 this work. However, the N present in OW cannot fertilize all the land available for corn
414 production in any of the studied scenarios. Consequently, strategies S1 and S2 must be
415 combined in the Pre-Directive scenario to maximize CI_C and CI_N . As Figure 5B shows, more
416 area is fertilized with the recovered products in the Pre-Directive scenario because of the higher
417 amount of OW that can be recycled, which makes farmers less dependent on industrial fertilizers
418 (strategy S3). Oppositely, the optimization of all the objective functions are partially based on
419 strategy S3 in the Post-Directive scenario.

420

421 The optimization of some objective functions provides duplicate or very similar results
422 (freshwater eutrophication and CI_P on the one hand, CI_C and different circularity indicators in
423 each scenario on the other). To avoid redundant results, freshwater eutrophication and CI_C
424 were not considered in the next part of the study, focused on a multi-objective optimization of
425 the other four objective functions.

426

427

428

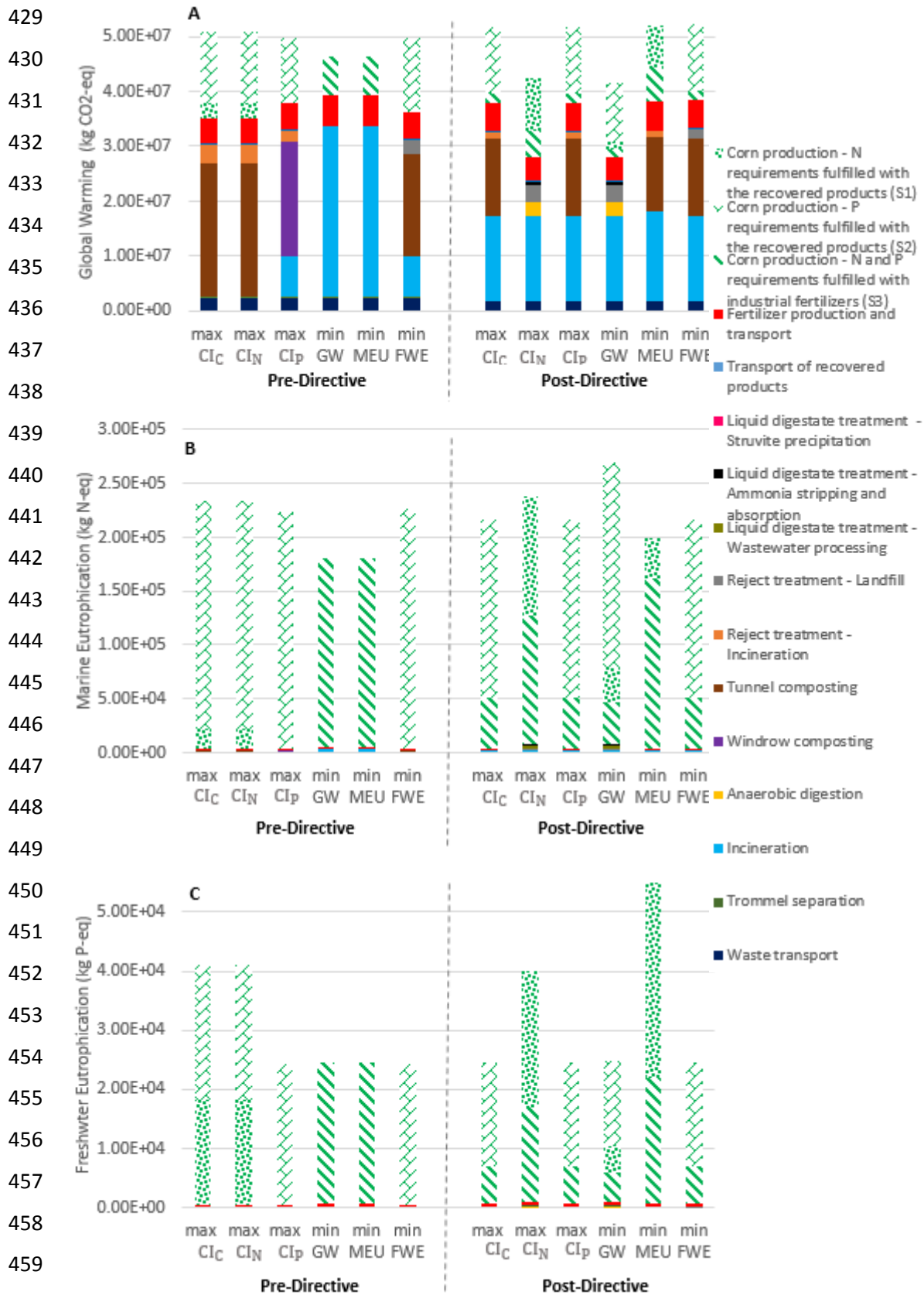


Figure 6. Contribution of the UPs to the environmental impacts in the Pre-Directive and Post-Directive scenarios

463

464 Figure 7 shows the Pareto fronts of the two scenarios, where each point is better than the others
465 in at least one of the values of the objective functions. Global warming and marine
466 eutrophication are normalized with respect to the maximum value of the two scenarios.

467

468 As the results of the DNDC simulations show, if industrial fertilizers, ammonium sulphate or
469 struvite (inorganic fertilizers) are exclusively applied to soil, the corn Nitrogen Use Efficiency
470 (NUE, defined as the fraction of N input harvested as product)⁵² decays over time because of the
471 depletion of SOC. The opposite occurs when bio-stabilized material, compost and digestate
472 (organic fertilizers) are applied, due to their C rich composition. However, the mean NUE
473 obtained for the 100-year time horizon if inorganic fertilizers are applied to land is higher than
474 the NUE achieved after the soil application of the organic fertilizers, because the share of plant
475 available inorganic N in the latter is low. This implies that more N leaches when the organic
476 fertilizers with a high organic N content are applied to land. These results are supported by
477 previous studies that highlight that the N leaching rate of organic fertilizers is higher than that of
478 inorganic fertilizers.^{53,54}

479

480 As Figures 6B and 6C indicate, the corn production subsystem is the main contributor to the
481 eutrophication impacts. In both scenarios the marine eutrophication impacts increase with the
482 CI_N , being the values of these two objective functions higher in the Post-Directive scenario. A
483 similar correlation cannot be established between CI_P and freshwater eutrophication because,
484 unlike N, which tends to leach as nitrate when it is applied to soil, P is strongly sorbed onto soil
485 particles; in fact its major environmental losses can be attributed to erosion.⁵⁵

486

487 The Pre-Directive scenario, where the minimum amount of OW that must be recycled is not
488 restricted, relies on incineration and the application of industrial fertilizers. Figure 6A shows

489 that, although the production of industrial fertilizers is very energy intensive,⁵⁶ the carbon
490 footprint associated with their land application is lower than that of the organic fertilizers, a
491 fraction of which degrades to CO₂ after their land application. Thus, as Figure 7A shows, in the
492 Pre-Directive scenario as CI_N increases, the CO₂-eq emissions increase too.

493

494 Anaerobic digestion is the UP that handles OW with the lowest carbon footprint. Hence, the
495 minimum carbon footprint achieved at the Post-Directive scenario, the only one where SS-OW
496 can be subjected to anaerobic digestion, is lower than in the Pre-Directive scenario. Moreover,
497 since the N recycling efficiency of anaerobic digestion and the LD UPs is higher than that of the
498 other UPs, the land application of the products derived from anaerobic digestion also maximizes
499 CI_N . Therefore, as shown in Figure 7B, in the Post-Directive scenario as the CI_N increases, the
500 carbon footprint of the system decreases.

501

502 Regarding CI_P , it shows a similar trend to the CI_N in the Pre-Directive scenario, whereas no clear
503 trend can be appreciated in the Post-Directive scenario, where the maximization of CI_P is
504 based on the application of compost to cover the soil P requirements, and the maximization of
505 CI_N on the application of ammonium sulphate and solid digestate to fulfill the soil N needs,
506 which leads to the accumulation of P in soil. The values of CI_P are lower in the Post-Directive
507 scenario because of the restriction that prevents mix-OW from being recycled.

508

509 A sensitivity analysis was performed to ascertain the consequences that a 20% decrease in the
510 values of two key parameters have on the results. The Spanish legislation prioritizes electricity
511 from the biogas produced at landfills and anaerobic digestion facilities over other sources of
512 non-renewable electricity. Notwithstanding, the electricity generated from waste incineration
513 does not have priority access to the grid.⁵⁷ The sensitivity analysis considered that 80% of the
514 electricity generated from the incineration of OW replaced the electricity from the Spanish grid

515 mix. On the other hand, it is hard to estimate the composition of SS-OW, since pilot experiments
516 for the source separation of OW have not been carried out in Cantabria. The sensitivity analysis
517 assumed that the fraction of OW in the SS-OW was 78.4%.

518

519 The results of the single-objective optimization of each scenario under the conditions of the
520 uncertainty analysis are compiled in Appendix E of the Supporting Information. The main
521 difference in the values of the decision variables after the performance of the sensitivity analysis
522 is that the freshwater eutrophication impacts of incineration exceed those of landfill. Thus,
523 landfill is selected over incineration when the freshwater eutrophication impacts are minimized.
524 As expected, the results of the sensitivity analysis led to slightly higher environmental impacts
525 in both scenarios and lower circularity indicators in the Post-Directive scenario.

526

527 Figure 7 proves that the environmental impacts associated with increasing the circularity of
528 nutrients cannot be overlooked. Whereas in the pre-Directive scenario there is a clear opposite
529 trend between the environmental impacts and the circularity of nutrients, the behavior of the
530 system in the Post-Directive scenario, subject to more restrictions and with more available UPs,
531 is more complex.

532

533 The findings of this study suggest that increasing the SSR of OW leads to a reduction in the
534 carbon footprint of the system. Although the results indicate that increasing the circularity of N
535 has detrimental eutrophication impacts, these are highly dependent on the sensitivity of the
536 receiving environment;⁵⁸ thus general conclusions cannot be drawn.

537

538 Before selecting a system configuration that meets the sustainability concerns and satisfies the
539 interests of all the stakeholders involved in waste management and the purchase of the
540 recovered products, a trade-off between the studied indicators must be identified. Moreover,

541 additional impact categories that quantify the environmental impacts associated with the
542 presence of heavy metals or organic pollutants in the recovered products, such as human
543 toxicity or ecotoxicity, should be included in the analysis. However, the feasibility of any system
544 configuration cannot be demonstrated until an economic analysis is performed.

545

546

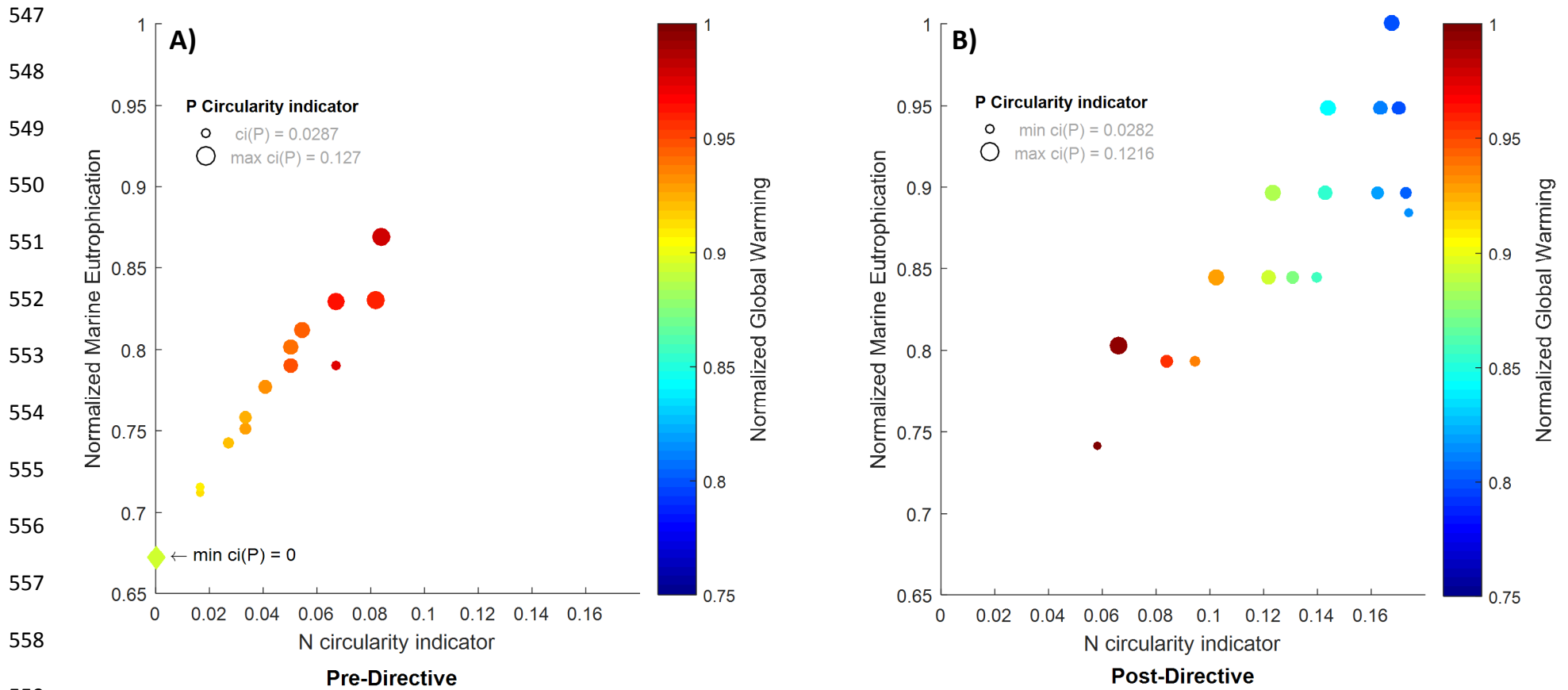


Figure 7. Pareto points for the Pre-Directive and Post-Directive scenarios

561 **NOMENCLATURE**

562

563 C – Carbon

564 CI_C – Carbon circularity indicator

565 CI_N – Nitrogen circularity indicator

566 CI_P – Phosphorus circularity indicator

567 CIWMS – Circular Integrated Waste Management System

568 DOC – Dissolved Organic Carbon

569 LCA – Life Cycle Assessment

570 LD – Liquid digestate

571 MFA – Material Flow Analysis

572 mix-OW – Organic waste separated from the mixed waste stream

573 N – Nitrogen

574 NUE – Nitrogen Use Efficiency

575 OW – Organic waste

576 P – Phosphorus

577 SOC – Soil Organic Carbon

578 SSR – Source Separation Rate

579 SS-OW – Source separated organic waste

580 UP – Unit Process

581

582

583 **SUPPORTING INFORMATION**

584

585 Waste composition, model data, sensitivity analysis.

586

587

588 **ACKNOWLEDGEMENTS**

589

590 The authors acknowledge the financial support from the Spanish MECD (FPU15/01771) and
591 MINECO (CTQ2016-76231-C2-1R).

592

593

594 **REFERENCES**

595

- 596 1. *Closing the loop - An EU action plan for the Circular Economy*. European Commission:
597 Brussels, 2015; [http://eur-lex.europa.eu/resource.html?uri=cellar:8a8ef5e8-99a0-](http://eur-lex.europa.eu/resource.html?uri=cellar:8a8ef5e8-99a0-11e5-b3b7-01aa75ed71a1.0012.02/DOC_1&format=PDF)
598 [11e5-b3b7-01aa75ed71a1.0012.02/DOC_1&format=PDF](http://eur-lex.europa.eu/resource.html?uri=cellar:8a8ef5e8-99a0-11e5-b3b7-01aa75ed71a1.0012.02/DOC_1&format=PDF).
- 599 2. Domenech, T.; Bahn-Walkowiak, B. Transition Towards a Resource Efficient Circular
600 Economy in Europe: Policy Lessons From the EU and the Member States. *Ecol.*
601 *Econ.* **2017**, DOI 10.1016/j.ecolecon.2017.11.001.
- 602 3. Delgado-Aguilar, M.; Tarrés, Q.; Pèlach, M. À.; Mutjé, P.; Fullana-I-Palmer, P. Are
603 Cellulose Nanofibers a Solution for a More Circular Economy of Paper Products? *Environ.*
604 *Sci. Technol.* **2015**, *49*, 12206-12213.
- 605 4. *The circularity gap report. An analysis of the circular state of the global economy*. Circle
606 economy: Amsterdam, 2018; <https://www.circularity-gap.world/report>.
- 607 5. Moriguchi, Y. Material flow indicators to measure progress toward a sound material-
608 cycle society. *J. Mater. Cycles Waste Manage.* **2007**, *9*, 112-120.
- 609 6. Haupt, M.; Vadenbo, C.; Hellweg, S. Do We Have the Right Performance Indicators for
610 the Circular Economy?: Insight into the Swiss Waste Management System. *J. Ind.*
611 *Ecol.* **2017**, *21*, 615-627.

- 612 7. Franklin-Johnson, E.; Figge, F.; Canning, L. Resource duration as a managerial indicator
613 for Circular Economy performance. *J. Clean. Prod.* **2016**, *133*, 589-598.
- 614 8. Circularity Indicators. An approach to measuring circularity. Methodology; Ellen
615 MacArthur Foundation, 2015;
616 <https://www.ellenmacarthurfoundation.org/programmes/insight/circularity-indicators>
- 617 9. Cobo, S.; Dominguez-Ramos, A.; Irabien, A. From linear to circular integrated waste
618 management systems: A review of methodological approaches. *Resour. Conserv.*
619 *Recycl.* **2017**, DOI 10.1016/j.resconrec.2017.08.003.
- 620 10. Directive on waste and repealing certain Directives. Directive 2008/98/EC, 2008;
621 <http://eur-lex.europa.eu/legal-content/En/TXT/?uri=celex%3A32008L0098>.
- 622 11. *Plan de residuos de la Comunidad Autónoma de Cantabria 2016 – 2022*; Gobierno de
623 Cantabria, Consejería de Universidades e Investigación, Medio Ambiente y Política
624 Social: Santander, 2016;
625 http://www.medioambientecantabria.es/documentos_contenidos/64293_2.versioninicialPR.pdf.
- 626
- 627 12. Cordell, D.; Drangert, J.-L.; White, S. The story of phosphorus: Global food security and
628 food for thought. *Global Environ. Change* **2009**, *19*, 292-305.
- 629 13. Rockström, J.; Steffen, W.; Noone, K.; Persson, A.; Chapin III, F. S.; Lambin, E.; Lenton, T.
630 M.; Scheffer, M.; Folke, C.; Schellnhuber, H. J.; Nykvist, B.; de Wit, C. A.; Hughes, T.; van
631 der Leeuw, S.; Rodhe, H.; Sörlin, S.; Snyder, P. K.; Costanza, R.; Svedin, U.; Falkenmark,
632 M.; Karlberg, L.; Corell, R. W.; Fabry, V. J.; Hansen, J.; Walker, B.; Liverman, D.;
633 Richardson, K.; Crutzen, P.; Foley, J. Planetary boundaries: Exploring the safe operating
634 space for humanity. *Ecol. Soc.* **2009**, *14*(2):32.
- 635 14. Likens, G.E.; Bormann, F.H.; Johnson, N.M. Interactions between major biogeochemical
636 cycles in terrestrial ecosystems. In *Some perspectives of the major biogeochemical*
637 *cycles*; Likens, G.E., Ed.; John Wiley & Sons: Chichester, 1981; pp 93-109.

- 638 15. Lal, R. Soil carbon sequestration impacts on global climate change and food
639 security. *Science* **2004**, *304*, 1623-1627.
- 640 16. Kroeze, C.; Hofstra, N.; Ivens, W.; Löhr, A.; Strokal, M.; van Wijnen, J. The links between
641 global carbon, water and nutrient cycles in an urbanizing world - the case of coastal
642 eutrophication. *Curr. Opin. Environ. Sustainability* **2013**, *5*, 566-572.
- 643 17. Hanserud, O. S.; Cherubini, F.; Øgaard, A. F.; Müller, D. B.; Brattebø, H. Choice of mineral
644 fertilizer substitution principle strongly influences LCA environmental benefits of
645 nutrient cycling in the agri-food system. *Sci. Total Environ.* **2018**, *615*, 219-227.
- 646 18. Schoumans, O.F. Phosphorus leaching from soils: process description, risk assessment
647 and mitigation. Ph.D. Dissertation, Wageningen University, Wageningen, The
648 Netherlands, 2015.
- 649 19. Rowe, H.; Withers, P. J. A.; Baas, P.; Chan, N. I.; Doody, D.; Holiman, J.; Jacobs, B.; Li, H.;
650 MacDonald, G. K.; McDowell, R.; Sharpley, A. N.; Shen, J.; Taheri, W.; Wallenstein, M.;
651 Weintraub, M. N. Integrating legacy soil phosphorus into sustainable nutrient
652 management strategies for future food, bioenergy and water security. *Nutr. Cycl.*
653 *Agroecosyst.* **2016**, *104*, 393-412.
- 654 20. Yao, Y.; Martinez-Hernandez, E.; Yang, A. Modelling nutrient flows in a simplified local
655 food-energy-water system. *Resour. Conserv. Recycl.* **2018**, DOI
656 10.1016/j.resconrec.2018.02.022.
- 657 21. Tonini, D.; Martinez-Sanchez, V.; Astrup, T. F. Material resources, energy, and nutrient
658 recovery from waste: Are waste refineries the solution for the future? *Environ. Sci.*
659 *Technol.* **2013**, *47*, 8962-8969.
- 660 22. Wang, X.; Guo, M.; Koppelaar, R. H. E. M.; Van Dam, K. H.; Triantafyllidis, C. P.; Shah, N.
661 A Nexus Approach for Sustainable Urban Energy-Water-Waste Systems Planning and
662 Operation. *Environ. Sci. Technol.* **2018**, *52*, 3257-3266.

- 663 23. Knoop, C.; Tietze, M.; Dornack, C.; Raab, T. Fate of nutrients and heavy metals during
664 two-stage digestion and aerobic post-treatment of municipal organic waste. *Bioresour.*
665 *Technol.* **2018**, *251*, 238-248.
- 666 24. Yoshida, H.; ten Hoeve, M.; Christensen, T. H.; Bruun, S.; Jensen, L. S.; Scheutz, C. Life
667 cycle assessment of sewage sludge management options including long-term impacts
668 after land application. *J. Clean. Prod.* **2018**, *174*, 538-547.
- 669 25. *Los pastos en Cantabria y su aprovechamiento. Memoria*; Centro de Investigación y
670 Formación Agrarias: Santander, 2006;
671 <http://cifacantabria.org/Documentos/memoria.pdf>.
- 672 26. Ariunbaatar, J.; Panico, A.; Esposito, G.; Pirozzi, F.; Lens, P. N. L. Pretreatment methods
673 to enhance anaerobic digestion of organic solid waste. *Appl. Energy* **2014**, *123*, 143-156.
- 674 27. Bernstad, A.; la Cour Jansen, J. Separate collection of household food waste for
675 anaerobic degradation - Comparison of different techniques from a systems
676 perspective. *Waste Manage.* **2012**, *32*, 806-815.
- 677 28. Bernstad, A.; Malmquist, L.; Truedsson, C.; la Cour Jansen, J. Need for improvements in
678 physical pretreatment of source-separated household food waste. *Waste*
679 *Manage.* **2013**, *33*, 746-754.
- 680 29. Carlsson, M.; Holmström, D.; Bohn, I.; Bisailon, M.; Morgan-Sagastume, F.; Lagerkvist,
681 A. Impact of physical pre-treatment of source-sorted organic fraction of municipal solid
682 waste on greenhouse-gas emissions and the economy in a Swedish anaerobic digestion
683 system. *Waste Manage.* **2015**, *38*, 117-125.
- 684 30. Carlsson, M.; Naroznova, I.; Moller, J.; Scheutz, C.; Lagerkvist, A. Importance of food
685 waste pre-treatment efficiency for global warming potential in life cycle assessment of
686 anaerobic digestion systems. *Resour. Conserv. Recycl.* **2015**, *102*, 58-66.

- 687 31. Cobo, S.; Dominguez-Ramos, A.; Irabien, A. Minimization of Resource Consumption and
688 Carbon Footprint of a Circular Organic Waste Valorization System. *ACS Sustainable*
689 *Chem. Eng.* **2018**, *6*, 3493-3501.
- 690 32. Steubing, B.; Mutel, C.; Suter, F.; Hellweg, S. Streamlining scenario analysis and
691 optimization of key choices in value chains using a modular LCA approach. *Int. J. Life*
692 *Cycle Assess.* **2016**, *21*, 510-522.
- 693 33. Haupt, M.; Kägi, T.; Hellweg, S. (2018). Modular life cycle assessment of municipal solid
694 waste management. *Waste Manage.* **2018**, DOI:10.1016/j.wasman.2018.03.035.
- 695 34. Clavreul, J.; Baumeister, H.; Christensen, T. H.; Damgaard, A. An environmental
696 assessment system for environmental technologies. *Environ. Model. Softw.* **2014**, *60*,
697 18-30.
- 698 35. Gilhespy, S. L.; Anthony, S.; Cardenas, L.; Chadwick, D.; del Prado, A.; Li, C.; Misselbrook,
699 T.; Rees, R. M.; Salas, W.; Sanz-Cobena, A.; Smith, P.; Tilston, E. L.; Topp, C. F. E.; Vetter,
700 S.; Yeluripati, J. B. First 20 years of DNDC (DeNitrification DeComposition): Model
701 evolution. *Ecol. Model.* **2014**, *292*, 51-62.
- 702 36. Lehmann, J.; Kleber, M. The contentious nature of soil organic matter. *Nature* **2015**, *528*,
703 60-68.
- 704 37. Foley, J. A.; DeFries, R.; Asner, G. P.; Barford, C.; Bonan, G.; Carpenter, S. R.; Chapin, F.
705 S.; Coe, M. T.; Daily, G. C.; Gibbs, H. K.; Helkowski, J. H.; Holloway, T.; Howard, E. A.;
706 Kucharik, C. J.; Monfreda, C.; Patz, J. A.; Prentice, I. C.; Ramankutty, N.; Snyder, P. K.
707 Global consequences of land use. *Science* **2005**, *309*, 570-574.
- 708 38. Gougoulias, C.; Clark, J. M.; Shaw, L. J. The role of soil microbes in the global carbon
709 cycle: Tracking the below-ground microbial processing of plant-derived carbon for
710 manipulating carbon dynamics in agricultural systems. *J. Sci. Food Agric.* **2014**, *94*, 2362-
711 2371.

- 712 39. *Encuesta sobre superficies y rendimientos cultivos*; Ministerio de Agricultura y Pesca,
713 Alimentación y Medio Ambiente: Madrid, 2017;
714 <http://www.mapama.gob.es/es/estadistica/temas/estadisticas->
715 [agrarias/agricultura/esyrce/](http://www.mapama.gob.es/es/estadistica/temas/estadisticas-agrarias/agricultura/esyrce/).
- 716 40. Council Directive on the landfill of waste. Council Directive 1999/31/EC,
717 1999;[http://eur-lex.europa.eu/legal-](http://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:31999L0031&from=EN)
718 [content/EN/TXT/PDF/?uri=CELEX:31999L0031&from=EN](http://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:31999L0031&from=EN).
- 719 41. *Source separation of MSW. An overview of the source separation and separate collection*
720 *of the digestible fraction of household waste, and of other similar wastes from*
721 *municipalities, aimed to be used as feedstock for anaerobic digestion in biogas plants*;
722 IEA bioenergy: 2013; [http://task37.ieabioenergy.com/files/daten-](http://task37.ieabioenergy.com/files/daten-redaktion/download/Technical%20Brochures/source_separation_web.pdf)
723 [redaktion/download/Technical%20Brochures/source_separation_web.pdf](http://task37.ieabioenergy.com/files/daten-redaktion/download/Technical%20Brochures/source_separation_web.pdf).
- 724 42. Copado-Méndez, P. J.; Pozo, C.; Guillén-Gosálbez, G.; Jiménez, L. Enhancing the ϵ -
725 constraint method through the use of objective reduction and random sequences:
726 Application to environmental problems. *Comput. Chem. Eng.* **2016**, *87*, 36-48.
- 727 43. Crenna, E.; Sozzo, S.; Sala, S. Natural biotic resources in LCA: Towards an impact
728 assessment model for sustainable supply chain management. *J. Clean. Prod.* **2018**, *172*,
729 3669-3684.
- 730 44. Notarnicola, B.; Sala, S.; Anton, A.; McLaren, S. J.; Saouter, E.; Sonesson, U. The role of
731 life cycle assessment in supporting sustainable agri-food systems: A review of the
732 challenges. *J. Clean. Prod.* **2017**, *140*, 399-409.
- 733 45. Frischknecht, R. Life cycle inventory analysis for decision-making. Scope-dependent
734 inventory system models and context-specific joint product allocation. Ph.D.
735 dissertation, Swiss Federal Institute of Technology, Zurich, 1998.

- 736 46. Li, C.; Frolking, S., Frolking, T. A. A model of nitrous oxide evolution from soil driven by
737 rainfall events: 1. model structure and sensitivity. *J. Geophys. Res.* **1992**, *97*(D9), 9759-
738 9776.
- 739 47. Frossard, E.; Skrabal, P.; Sinaj, S.; Bangerter, F.; Traore, O. Forms and exchangeability of
740 inorganic phosphate in composted solid organic wastes. *Nutr. Cycl. Agroecosyst.* **2002**,
741 *62*, 103-113.
- 742 48. Gagnon, B.; Demers, I.; Ziadi, N.; Chantigny, M. H.; Parent, L. -.; Forge, T. A.; Larney, F.
743 J.; Buckley, K. E. Forms of phosphorus in composts and in compostamended soils
744 following incubation. *Can. J. Soil Sci.* **2012**, *92*, 711-721.
- 745 49. García-Albacete, M.; Martín, A.; Cartagena, M. C. Fractionation of phosphorus
746 biowastes: Characterisation and environmental risk. *Waste Manage.* **2012**, *32*, 1061-
747 1068.
- 748 50. Hansen, T. L.; Bhandar, G. S.; Christensen, T. H.; Bruun, S.; Jensen, L. S. Life cycle
749 modelling of environmental impacts of application of processed organic municipal solid
750 waste on agricultural land (Easewaste). *Waste Manage. Res.* **2006**, *24*, 153-166.
- 751 51. Tampio, E.; Marttinen, S.; Rintala, J. Liquid fertilizer products from anaerobic digestion
752 of food waste: Mass, nutrient and energy balance of four digestate liquid treatment
753 systems. *J. Clean. Prod.* **2016**, *125*, 22-32.
- 754 52. Zhang, X.; Davidson, E. A.; Mauzerall, D. L.; Searchinger, T. D.; Dumas, P.; Shen, Y.
755 Managing nitrogen for sustainable development. *Nature* **2015**, *528*, 51-59.
- 756 53. Yoshida, H.; Nielsen, M. P.; Scheutz, C.; Jensen, L. S.; Bruun, S.; Christensen, T. H. Long-
757 Term Emission Factors for Land Application of Treated Organic Municipal
758 Waste. *Environ. Model. Assess.* **2016**, *21*, 111-124.
- 759 54. Brockmann, D.; Pradel, M.; Hélias, A. Agricultural use of organic residues in life cycle
760 assessment: Current practices and proposal for the computation of field emissions and
761 of the nitrogen mineral fertilizer equivalent. *Resour. Conserv. Recycl.* **2018**, *133*, 50-62.

- 762 55. Esculier, F.; Le Noë, J.; Barles, S.; Billen, G.; Créno, B.; Garnier, J.; Lesavre, J.; Petit, L.;
763 Tabuchi, J. P. The biogeochemical imprint of human metabolism in Paris Megacity: A
764 regionalized analysis of a water-agro-food system. *J. Hydrol.* **2018**,
765 DOI:10.1016/j.jhydrol.2018.02.043.
- 766 56. Snyder, C. S.; Bruulsema, T. W.; Jensen, T. L.; Fixen, P. E. Review of greenhouse gas
767 emissions from crop production systems and fertilizer management effects. *Agric.*
768 *Ecosyst. Environ.* **2009**, *133*, 247-266.
- 769 57. Real Decreto por el que se regula la actividad de producción de energía eléctrica a partir
770 de fuentes de energía renovables, cogeneración y residuos. Real Decreto 413/2014,
771 2014; https://www.boe.es/diario_boe/txt.php?id=BOE-A-2014-6123.
- 772 58. De Jonge, V. N.; Elliott, M.; Orive, E. Causes, historical development, effects and future
773 challenges of a common environmental problem: Eutrophication. *Hydrobiologia* **2002**,
774 *475-476*, 1-19.
- 775